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# Scanning tunneling microscopy investigation of single electron tunneling in Co–Al–O and Cu–Al–O granular films

K. Takanashi,[a] S. Mitani, J. Chiba, and H. Fujimori
*Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan*

We have investigated single electron tunneling in Co–Al–O and Cu–Al–O granular films using scanning tunneling microscopy (STM). Topographic images show well-defined granular structures where nanometer-sized metal granules are embedded in insulating matrix. The Coulomb staircases in the current–voltage ($I-V$) curves are clearly observed even at room temperature in both films. For the Co–Al–O film, furthermore, negative differential conductance appears in the Coulomb staircase. © *2000 American Institute of Physics.* [S0021-8979(00)74808-0]

## I. INTRODUCTION

Spin dependent tunneling transport leading to giant magnetoresistance (GMR) has attracted much attention in recent years because of both the fundamental physics behind the phenomena and the potential for possible application.[1] Not only planar-type junctions, but also insulating granular systems such as Co–Al–O films,[2] where magnetic metal granules (e.g., Co) are embedded in insulating matrix (e.g., Al–oxide), are known to show tunnel GMR. In granular systems, the granules are so small (usually a few nanometers in diameter) that the Coulomb charging energy of the granules is significant enough to suppress the tunneling transport. This is called the Coulomb blockade. The Coulomb blockade is expected to have a significant influence on spin dependent tunneling. The tunnel GMR in granular systems was reported[3] to show enhancement at low temperatures due to the higher-order tunneling associated with Coulomb blockade. However, the transport in granular systems represents the statistical average of tunneling between many granules with various sizes and intergranular distances. In order to elucidate the interplay of Coulomb blockade and spin dependent tunneling, it is important to investigate the single electron tunneling between granules.

Scanning tunneling microscopy (STM) is a useful tool for the investigation of single electron tunneling. We reported preliminary results of the STM observation of a Co–Al–O granular film in a previous paper,[4] suggesting the appearance of Coulomb staircase structure in the current–voltage ($I-V$) curves even at room temperature, and Imamura *et al.*[5] gave a theoretical insight into the appearance of the Coulomb staircase. In this paper, we present the experimental results of STM topological images and $I-V$ measurements in Co–Al–O and Cu–Al–O granular films, to make a comparison between single electron phenomena in magnetic (Co) and nonmagnetic (Cu) granules.

## II. EXPERIMENTAL PROCEDURE

Samples were prepared on glass substrates by rf sputtering. For a Co–Al–O granular film, reactive sputtering was carried out using a Co–Al alloy target and an $Ar+O_2$ mixed gas. For a Cu–Al–O granular film, on the other hand, sputtering with pure Ar gas was carried out using a composite target: Cu sheets were placed on a $Al_2O_3$ target. The thicknesses of Co–Al–O and Cu–Al–O granular films were approximately 1 $\mu$m and 0.26 $\mu$m, respectively. The chemical compositions of films were determined to be $Co_{36}Al_{22}O_{42}$ and $Cu_{24}Al_{30}O_{46}$ by Rutherford backscattering spectrometry (RBS). For both films, the temperature ($T$) dependence of electrical resistivity $\rho$ gave the relationship $\ln \rho \sim T^{-1/2}$, indicating that the transport is dominated by tunneling of electrons between granules.[6] Furthermore, the $Co_{36}Al_{22}O_{42}$ film showed GMR of 10% at room temperature. The detailed behavior of electrical transport and GMR was described elsewhere.[3,7]

A STM apparatus (DME, Rasterscope 3000) was operated at room temperature under high vacuum ($\sim 10^{-8}$ Torr) using a Pt tip. The sample was mounted onto a copper plate, and silver paste was used to provide electrical conduction between the sample and the plate. Topographic images were obtained in a constant current mode with a negative bias voltage applied to the tip. For the $I-V$ measurements, the tip bias voltage was varied in the range of $-2.0$ to $+2.0$ V with a constant tip-to-sample distance.

## III. RESULTS AND DISCUSSION

Figures 1(a) and (b) show the topographic images for $Co_{36}Al_{22}O_{42}$ and $Cu_{24}Al_{30}O_{46}$ films, respectively. In both films, we observe similar granular structures consisting of round shaped bright regions and dark channels, which are associated with Co(Cu) granules and Al–oxide, respectively. The size of granules and the distance between the centers of two adjacent granules are estimated to be approximately 2–3 and 3–4 nm, respectively, in average. This result is consistent with the TEM observation[8] and the analysis of superparamagnetic magnetization curves[9] in the $Co_{36}Al_{22}O_{42}$ film.

Figures 2(a) and 2(b) show the typical $I-V$ curves for $Co_{36}Al_{22}O_{42}$ and $Cu_{24}Al_{30}O_{46}$ films, respectively. The measurements were made with the STM tip placed on a Co(Cu) granule. In both films, the tunnel current varies stepwise as a function of the applied voltage, indicating the appearance of Coulomb staircase. We have made many $I-V$ measurements with different tips and different portions of the sample. We always observed the clear reproducible Coulomb staircase

[a]Corresponding author: koki@imr.tohoku.ac.jp

 

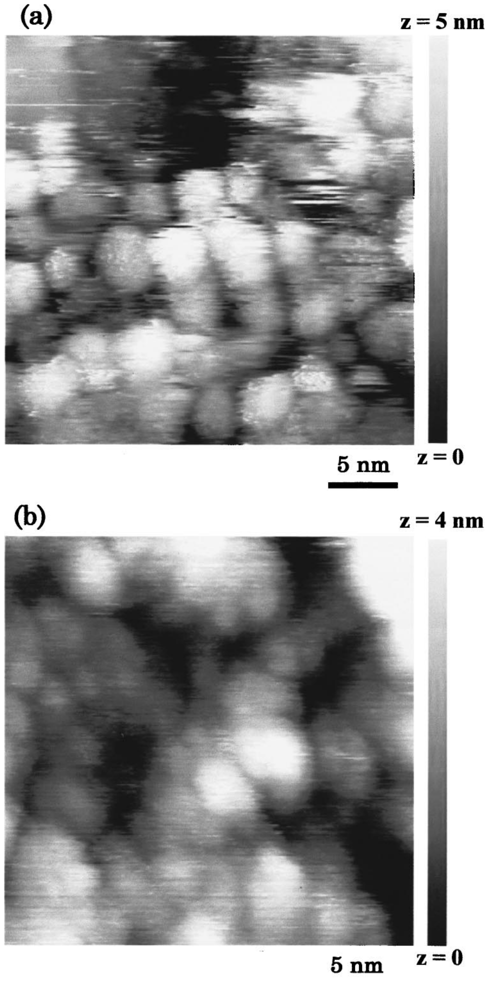


FIG. 1. STM topographic images of (a) $Co_{36}Al_{22}O_{42}$ and (b) $Cu_{24}Al_{30}O_{46}$ films.

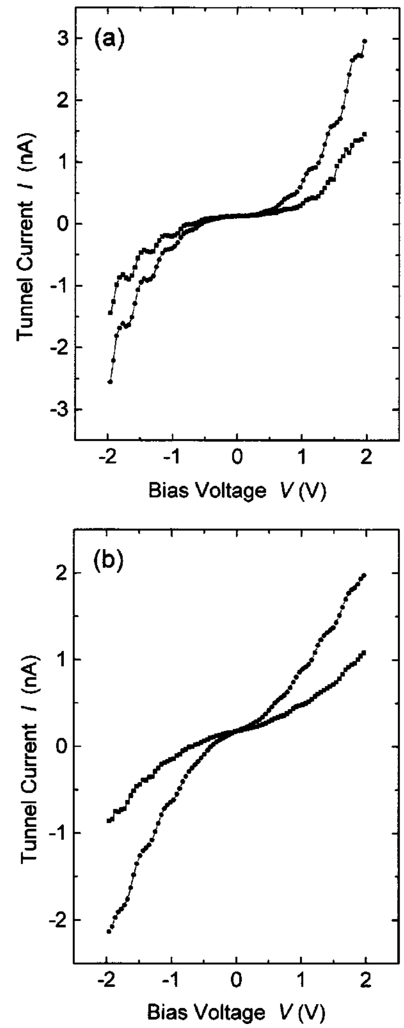


FIG. 2. Current vs applied voltage ($I-V$) curves measured at room temperature for (a) $Co_{36}Al_{22}O_{42}$ and (b) $Cu_{24}Al_{30}O_{46}$ films. The different two curves in each figure were obtained for different lateral positions.

structure, although the period of Coulomb staircase varies slightly with the position of the tip. Therefore, we believe that this Coulomb staircase structure is associated with the single electron tunneling from the tip through the granular film. The period of the Coulomb staircase is in the range of approximately 0.25–0.4 V. The capacitance of a completely isolated granule with the diameter $d=2-3$ nm, $2\pi\varepsilon_0 d$, ($\varepsilon_0$: the dielectric constant of vacuum), is estimated to be $1-2 \times 10^{-19}$ F, and the charging energy, $e^2/4\pi\varepsilon_0 d$, is 0.5–0.7 eV. The capacitance between the tip and a granule on the surface is somewhat larger than that of an isolated granule, and the charging energy may be reduced by a factor, considering that the tip is sufficiently large and the distance between the tip and the granule is of the order of 1 nm. Therefore, the period of Coulomb staircase is consistent with the capacitance between the tip and a granule on the surface.

In the present experiment, the thicknesses of granular films are so large compared to the size of granules and the intergranular distance, and there are a large number of granules in the conduction path from the tip through the film to the copper plate. The capacitances between granules in the film are much larger than those between the tip and a granule on the surface, and the charging energy is much smaller, because the insulating matrix (Al–oxide) has a large dielectric constant (approximately 10 times larger than that of vacuum) and a granule in the film are surrounded with several granules. The analysis of the $\ln\rho \sim T^{-1/2}$ curve for the $Co_{36}Al_{22}O_{42}$ film gave the charging energy of the order of 0.01 eV.[3] In the STM experiment, however, the resistance between the tip and a granule on the surface is $\sim$G$\Omega$; on the other hand, the resistances between granules in the film are $\sim$M$\Omega$ which is estimated from the value of $\rho$.[3,7] In other words, the bottleneck is formed between the tip and a granule on the surface. Then, the period of the Coulomb staircase should be dominated by the capacitance at the bottleneck, irrespective of the number of granules in the conduction path. We have also made the STM experiment of a 10 nm thick $Co_{36}Al_{22}O_{42}$ granular film with a CoAl alloy layer inserted between the granular film and the glass substrate as a base electrode. In this case, there are only 2 or 3 granules in the conduction path. We have also observed the Coulomb staircase structure in the $I-V$ curves for the 10 nm thick $Co_{36}Al_{22}O_{42}$ film, which is similar to that for the 1 $\mu$m thick film. Furthermore, the bottleneck between the tip and a granule on the surface is so narrow (the resistance is about three orders of magnitude larger than those between granules in the film) that the Coulomb staircase is not smeared out but clearly observed even at room temperature.[5]

Comparing the results for the $Co_{36}Al_{22}O_{42}$ film with those for the $Cu_{24}Al_{30}O_{46}$ film, there is no remarkable difference observed in the topographic image and in the period of the Coulomb staircase. For the $Co_{36}Al_{22}O_{42}$ film, however, negative differential conductance, i.e., $dI/dV<0$, appears on several steps of the Coulomb staircase in $I-V$ curves. The negative $dI/dV$ is reproducible on the same measurement condition, and definitely distinguishable from noise. For the $Cu_{24}Al_{30}O_{46}$ film, on the other hand, negative $dI/dV$ is not clearly observed. Barnas and Fert[10] predicted the appearance of negative $dI/dV$ due to spin accumulation effect, which might be a possible reason for the difference between the $I-V$ curves for $Co_{36}Al_{22}O_{42}$ and $Cu_{24}Al_{30}O_{46}$ films. In order to elucidate the spin contribution to single electron tunneling, more systematic studies including the temperature and magnetic field dependence of $I-V$ curves are now in progress.

In summary, we have made the STM investigation of Co–Al–O and Cu–Al–O granular films. Topographic images indicate well defined granular structures. We have observed Coulomb staircases in the $I-V$ curves at room temperature, which are associated with single electron tunneling from the tip through the granular film. For the Co–Al–O film, furthermore, negative $dI/dV$ is observed in the Coulomb staircase, suggesting spin accumulation effect.

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